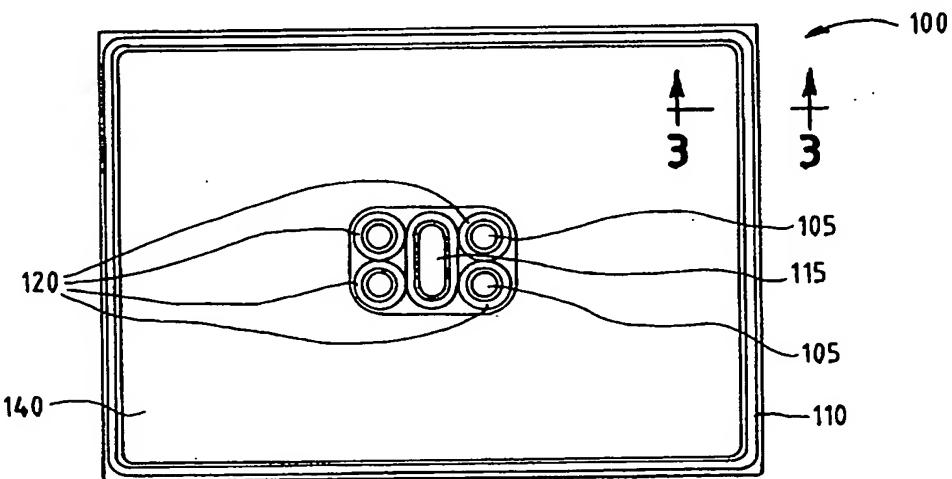




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(54) Title: RESILIENT SEAL FOR MEMBRANE ELECTRODE ASSEMBLY (MEA) IN AN ELECTROCHEMICAL FUEL CELL AND METHOD OF MAKING SAME



(57) Abstract

An improved membrane electrode assembly ("MEA") for an electrochemical fuel cell comprises coextensive ion exchange membrane and electrode layers and a resilient fluid impermeable integral seal comprising a sealant material impregnated into the porous electrode layers in sealing regions of the MEA. In preferred embodiments, the integral seal circumscribes the electrochemically active area of the MEA. The integral seal preferably also extends laterally beyond the edge of the MEA, thereby enveloping the peripheral region including the side edge of the MEA. An integral seal may also be provided around any openings, such as manifold openings, that are formed in the MEA. Preferably the sealant material is a flow processable elastomer applied to the MEA using an injection molding process prior to being cured. In preferred embodiments the seal has a plurality of spaced parallel raised ribs with cross ribs extending therebetween at spaced intervals. The raised ribs and cross ribs provide compartmentalized seals that provide improved protection against fluid leaks in a fuel cell assembly.

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RESILIENT SEAL FOR MEMBRANE ELECTRODE ASSEMBLY (MEA) IN AN ELECTROCHEMICAL FUEL CELL AND METHOD OF MAKING SAME

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Technical Field

The present invention relates to electrochemical fuel cells. In particular, the invention provides an improved membrane electrode assembly for a fuel cell, and a method of making an improved membrane electrode assembly. An improved membrane electrode assembly comprises integral fluid impermeable seals and coextensive electrode and membrane layers.

Background

15 Electrochemical fuel cells convert reactants, namely fuel and oxidant fluid streams, to generate electric power and reaction products. Electrochemical fuel cells employ an electrolyte disposed between two electrodes, namely a cathode and an anode. The electrodes each comprise an electrocatalyst disposed at the interface between the electrolyte and the electrodes to induce the desired electrochemical reactions. The location of the 20 electrocatalyst generally defines the electrochemically active area.

25 Solid polymer fuel cells generally employ a membrane electrode assembly (“MEA”) consisting of a solid polymer electrolyte or ion exchange membrane disposed between two electrode layers comprising porous, electrically conductive sheet material. The membrane is ion conductive (typically proton conductive), and also acts as a barrier for isolating the reactant streams from each other. Another function of the membrane is to act 30 as an electrical insulator between the two electrode layers. The electrodes must be electrically insulated from each other to prevent short-circuiting. If a multi-layer MEA is cut, tiny portions of the electrically conductive electrode material, such as stray fibers, may bridge across the thin membrane, interconnecting the electrodes, which could cause electrical short-circuiting in an operating fuel cell. Conventional MEAs incorporate a

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membrane with a larger surface area than the electrode layers, with at least a small portion of the membrane extending laterally beyond the edge of the electrode layers. The protruding membrane edge helps to prevent short-circuiting between the electrodes around the edge of the membrane. A problem with this is that it is difficult to cut an MEA after the electrodes have been joined to the membrane so that the thin membrane has a larger area than the electrodes. A conventional MEA is fabricated by manufacturing and cutting the electrodes and membrane layers separately. After the electrodes and membrane have been cut to the desired size and shape, the cut electrode layers are laminated with the cut membrane layer. These steps are not conducive to high speed manufacturing processes. It would be preferable to manufacture a sheet or roll of MEA material that already comprises the electrode and membrane layers, wherein this multi-layer material could then be cut to the desired size and shape for individual MEAs. An MEA cut in this way, such that the electrodes and membrane are coextensive, is described herein as being a "flush cut" MEA. However, this approach has heretofore been impractical because of the short circuiting problem described above.

In a fuel cell stack, the MEA is typically interposed between two separator plates that are substantially impermeable to the reactant fluid streams. The plates act as current collectors and provide support for the electrodes. To control the distribution of the reactant fluid streams to the electrochemically active area, the surfaces of the plates that face the MEA may have open-faced channels or grooves formed therein. Such channels or grooves define a flow field area that generally corresponds to the adjacent electrochemically active area. Such separator plates, which have reactant channels formed therein are commonly known as flow field plates. In a fuel cell stack a plurality of fuel cells are connected together, typically in series, to increase the overall output power of the assembly. In such an arrangement, one side of a given plate may serve as an anode plate for one cell and the other side of the plate may serve as the cathode plate for the adjacent cell. In this arrangement the plates may be referred to as bipolar plates.

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The fuel fluid stream that is supplied to the anode typically comprises hydrogen. For example, the fuel fluid stream may be a gas such as substantially pure hydrogen or a reformate stream containing hydrogen. Alternatively, a liquid fuel stream such as aqueous methanol may be used. The oxidant fluid stream, which is supplied to the cathode, typically 5 comprises oxygen, such as substantially pure oxygen, or a dilute oxygen stream such as air. In a fuel cell stack, the reactant streams are typically supplied and exhausted by respective supply and exhaust manifolds. Manifold ports are provided to fluidly connect the manifolds to the flow field area and electrodes. Manifolds and corresponding ports may also be provided for circulating a coolant fluid through interior passages within the stack to absorb 10 heat generated by the exothermic fuel cell reactions.

It is desirable to seal reactant fluid stream passages to prevent leaks or inter-mixing of the fuel and oxidant fluid streams. Fuel cell stacks typically employ resilient seals between stack components. Such seals isolate the manifolds and the electrochemically active area of the fuel cell MEAs by circumscribing these areas. For example, a fluid tight 15 seal may be achieved in a conventional fuel cell stack by using elastomeric gasket seals interposed between the flow field plates and the membrane, with sealing effected by applying a compressive force to the resilient gasket. Accordingly, it is important for conventional fuel cell stacks to be equipped with seals and a suitable compression assembly for applying a compressive force to the seals.

20 Conventional methods of sealing around plate manifold openings and MEAs within fuel cells include framing the MEA with a resilient fluid impermeable gasket, placing preformed gaskets in channels in the electrode layers and/or separator plates, or molding seals within grooves in the electrode layer or separator plate, circumscribing the electrochemically active area and any fluid manifold openings. Examples of conventional 25 methods are disclosed in U.S. Patent Nos. 5,176,966 and 5,284,718. Typically the gasket seals are cut from a sheet of gasket material. For a gasket seal that seals around the electrochemically active area of the MEA, the central portion of the sheet is cut away. This procedure results in a large amount of the gasket material being wasted. Because the

electrodes are porous, for the gasket seals to operate effectively, the gasket seals ordinarily are in direct contact with the flow field plates and the ion exchange membrane. Therefore, in a conventional MEA, electrode material is cut away in the sealing regions so that the gasket will contact the ion exchange membrane. Some MEAs use additional thin-film 5 layers to protect the ion exchange membrane where it would otherwise be exposed in the gasket seal areas. Separate components such as gasket seals and thin-film layers require respective processing or assembly steps, which add to the complexity and expense of manufacturing fuel cell stacks.

Accordingly, it is desirable to simplify and reduce the number of individual or 10 separate components involved in sealing in a fuel cell stack since this reduces assembly time and the cost of manufacturing.

Summary of the Invention

An improved MEA for an electrochemical fuel cell comprises:

15 a first porous electrode layer;
 a second porous electrode layer;
 an ion exchange membrane interposed between the first and second porous electrode layers wherein said first and second electrode layers and said membrane are coextensive;
20 electrocatalyst disposed at the interface between the ion exchange membrane and each of the first and second porous electrode layers, defining an electrochemically active area; and
 a resilient fluid impermeable seal integral with the MEA, comprising a fluid impermeable sealant material impregnated into the first and second porous electrode 25 layers in sealing regions thereof.

In a preferred embodiment the sealing regions comprise regions that circumscribe the electrochemically active area of the electrode layers. Preferably the sealant material impregnates a portion of the MEA electrodes in the peripheral region and extends laterally

beyond the edges of the electrode layers and membrane, (i.e. the sealant material envelops the membrane edge).

If the MEA further comprises one or more openings formed therein, such as opening for a fluid manifold and/or a tension member, the sealing region further may comprises 5 regions that circumscribe such openings.

In a fuel cell stack the sealing regions cooperate with the fuel cell separator plates to prevent fluids from leaking around the edges of the MEA. The sealant material is preferably an elastomer. In a preferred method of making an improved MEA, the sealant material is injection molded. Accordingly, it is desirable for the uncured sealant material to 10 be flow processable. After the uncured sealant material has been applied to the MEA, it is allowed to cure to form a resilient elastomeric material. The elastomeric sealant material may be a thermosetting material, as long as the curing temperature is compatible with the MEA components, and in particular, the ion exchange membrane.

The sealant material may also be used to form a reference feature such as a raised 15 edge or protrusion for assisting in the assembly of the fuel cell. For example, when an outer perimeter edge seal is being molded, at least one of the edges could be molded with a reference edge, which can be used to align the MEA during manufacturing processes. Alternatively, a protrusion such as a cylindrical plug could be molded in a location, which 20 can be aligned with a corresponding cylindrical depression in an adjacent separator plate during stack assembly.

From a manufacturing perspective, the coextensive electrode and membrane of the improved MEA provides advantages for high-speed manufacturing. For example, the electrode and membrane layers of the MEA may be formed in continuous processes that produce a multi-layered roll of material or large sheets that can be cut down to the size of an 25 individual MEA. This is difficult in conventional MEAs where the electrodes are not coextensive with the membrane (i.e. because the membrane extends laterally beyond the edge of the electrodes). However, because the electrodes and membrane are coextensive, the improved MEAs may be "flush cut" from a larger piece of multi-layered material.

An improved MEA with coextensive layers has been successfully manufactured by flush-cutting without short circuiting problems arising. Preferred aspects of the manufacturing process that have contributed to this success include one or more of the following:

- 5 (a) after flush cutting the multi-layer material, brushing the edges of the flush cut material to remove tiny portions of the electrode layer that may extend laterally beyond the edge of the membrane;
- (b) using a vacuum during the impregnation or injection molding process;
- (c) enveloping sealing regions of the MEA with a sealant material; and
- 10 (d) forming the integral seal and then flush cutting in the sealing regions.

It is believed that employing at least one of these approaches in the method of manufacturing an improved MEA provides significant benefits that help to prevent short circuiting caused by tiny particles of the electrodes straddling the membrane. For example, brushing the edges of the flush cut material may result in the removal of portions of the 15 electrode layer that extend laterally beyond the edge of the membrane. Applying a vacuum to the outer surfaces of the porous electrode layers during the impregnation or injection molding process helps to direct the flow of the sealant material so as to pull the electrode material away from the membrane and the opposing electrode while the sealant material is being applied. Another advantage of applying a vacuum is that it helps to remove air from 20 the mold and reduce the effect of bubble formation or foaming in the sealant material.

Furthermore, short circuiting may be reduced by enveloping peripheral regions of the MEA with an electrically insulating sealant material. The sealant material embeds the cut edge of the MEA so that the edges of the electrode layers are electrically insulated. The embedded electrode material is also immobilized by the sealant material so that the edges of the 25 electrodes can not be displaced by fluid currents or pressures within an operating fuel cell.

An additional benefit of enveloping peripheral regions of the MEA with the integral seal is that the fluid impermeable sealant material prevents dehydration of the membrane through the side edge.

In preferred embodiments, the integral seal further comprises a raised rib that is compressible when the MEA is placed between opposing fuel cell plates in an assembled fuel cell. The raised rib may cooperate with a recessed groove in the plate that provides a recessed sealing surface. The advantage of using a recessed sealing surface is that it is less susceptible to being damaged because the sealing surface is somewhat protected by being recessed. For example, a recessed sealing surface is less likely to be scratched when fuel cell plates are stacked on top of each other while being fabricated. Scratches in the sealing surface could cause leaks in an operating fuel cell. Ribs may be located in sealing regions where the sealant material is impregnated into the porous electrode layers (i.e. superposing the membrane) and/or may be located in regions where the sealant material extends laterally beyond the edges of the electrode layers and membrane.

10 In preferred embodiments, the integral seal of the MEA comprises a plurality of the raised ribs. For example, the ribs in each sealing region may be parallel, with each individual rib circumscribing the active area or the openings in the MEA. An advantage of having a plurality of raised ribs is increased protection against leaks. Each one of the plurality of ribs must be breached for there to be a fluid leak.

15 Versions of these preferred embodiments also employ raised cross-ribs between adjacent ones of the plurality of raised ribs. The cross-ribs compartmentalize the spaces between the raised ribs. Therefore, for there to be a leak, there must be a breach in the raised ribs adjoining the same sealed compartment; otherwise, any fluid leaking through a breach in a raised rib will be confined to the sealed compartment.

A method of making a MEA with resilient integral seals for use in an electrochemical fuel cell comprises the following sequential steps:

- (a) placing the MEA inside a mold;
- 25 (b) introducing a curable flow processable sealant material into the mold;
- (c) directing the sealant material to desired sealing regions of the MEA and impregnating a portion of an electrode layer of the MEA with the sealant material in the sealing regions; and

- (d) curing the sealant material to form an integral seal; and
- (e) removing the MEA from the mold.

In a preferred method step (c) is accompanied by the step of applying a vacuum to assist the injection and impregnation of the sealant material. If the vacuum is applied to 5 both major surfaces of the MEA, the vacuum also helps to prevent electrode material from bridging between the electrodes during the injection process.

A preferred method comprises using a sealant material that is a thermosetting material. This embodiment of the method further comprises the step of applying heat until the sealant material is cured. To cure the sealant material while it is still within the mold, 10 the temperature must be controlled to prevent overheating that may damage the MEA, and in particular the ion exchange membrane.

The mold preferably has open channels formed in the molding surface. The channels facilitate the distribution of the curable flow processable sealant material to the sealing regions. The channels also act as molding surfaces for forming ribs or ridges in the 15 integral seal. The mold is also preferably fitted with a raised dike. The dike impinges upon the MEA to limit the extent to which the sealant material impregnates the electrode layer. For example, the mold may have opposing dikes on opposite surfaces of the mold. When the mold is closed, the dikes press against and compress the electrode layers. The 20 compressed electrode layers have reduced porosity, which helps to confine the impregnation of the electrode layers to the sealing regions. The dikes are thus positioned on the sides of the sealing regions that face the electrochemically active areas of the MEA.

Brief Description Of The Drawings

The advantages, nature and additional features of the invention will become more 25 apparent from the following description, together with the accompanying drawings, in which:

FIG. 1 is a partially exploded isometric view of an electrochemical solid polymer fuel cell stack that incorporates an embodiment of an improved membrane electrode assembly that has integral fluid impermeable seals;

FIG. 2 is a plan view of an improved membrane electrode assembly;

5 FIGS. 3A through 3D are partial section views of an edge portion of the membrane electrode assembly of FIG. 2, as indicated by the section marked in FIG. 2;

FIGS. 4A through 4C are partial section views of the edges of three embodiments of a membrane electrode assembly interposed between two fuel cell separator plates with integral seals compressed therebetween; and

10 FIG. 5 is an enlarged plan view of a portion of a preferred embodiment of a membrane electrode assembly that has an integral seal that incorporates a plurality of seal ridges and cross-ridges.

Detailed Description of the Preferred Embodiments

15 FIG. 1 illustrates a solid polymer electrochemical fuel cell stack 10, including a pair of end plate assemblies 20 and 30, and a plurality of stacked fuel cell assemblies 50, each comprising an MEA 100, and a pair of flow field plates 200. A tension member 60 extends between end plate assemblies 20 and 30 to retain and secure stack 10 in its assembled state. Spring 70 with clamping members 80 grip each end of tension member 60 to apply a 20 compressive force to fuel cell assemblies 50 of stack 10.

Fluid reactant streams are supplied to and exhausted from internal manifolds and passages in stack 10 via inlet and outlet ports 40 in end plate assemblies 20 and 30. Aligned openings 105 and 205 in MEAs 100 and flow field plates 200, respectively, form reactant manifolds extending through stack 10.

25 In the illustrated embodiment, an integral perimeter seal 110 is provided around the outer edge of MEA 100. Integral manifold seals 120 circumscribe manifold openings 105. When stack 10 is secured in its assembled, compressed state, integral seals 110 and 120 cooperate with the adjacent pair of plates 200 to fluidly isolate fuel and oxidant reactant

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streams in internal reactant manifolds and passages, thereby isolating one reactant stream from the other and preventing the reactant streams from leaking from stack 10.

As illustrated in FIG. 1, each MEA 100 is positioned between the active surfaces of two flow field plates 200. Each flow field plate 200 has flow field channels 210 on the active surface thereof (which contacts the MEA) for distributing fuel or oxidant fluid streams to the active area of the MEA 100. In the embodiment illustrated in FIG. 1, flow field channels 210 are fluidly connected to manifold openings 205 in plate 200 via supply/exhaust channels 220 (partially shown) located on the non-active surface of flow field plate 200 and ports 230 extending through plate 200.

In the illustrated embodiment, flow field plates 200 have a plurality of open-faced parallel channels 250 formed in the non-active surface thereof. Channels 250 on adjacent pairs of plates 200 cooperate to form passages extending through stack 10, through which a coolant stream, such as air, may be directed.

FIG. 2 shows an MEA 100 with integral seals 110, 120 that respectively circumscribe the electrochemically active area of MEA 100, and manifold openings 105 and opening 115 through which tension member 60 extends. MEA 100 comprises an ion exchange membrane (not visible in FIG. 2) disposed between two porous, electrically conductive electrode layers 140. These electrode layers 140 may, for example be carbon fiber paper. A sealant material, preferably a flow processable elastomer, such as, for example, a thermosetting liquid injection moldable compound (e.g. silicones, fluoroelastomers, fluorosilicones, ethylene propylene di-methyl, and natural rubber), is impregnated into the porous electrode layers of MEA 100 to form integral seals 110 and 120.

Various embodiments of an MEA 100 with an integral seal such as 110, are illustrated in cross-sectional views in FIGS. 3A through 3D. The figures depict a perimeter edge integral seal 110, such as through section 3-3 of FIG. 2, although the same configurations could also be employed for integral seal 120 at a manifold opening (as in FIG. 1). Each embodiment of an MEA 100 comprises an ion exchange membrane 130

disposed between two porous, electrically conductive electrode layers 140, and a sealant material 125 impregnated into a portion 150 of the porous electrode layers of MEA 100. Preferably, at least a portion of seal 110 protrudes above the outer surface of porous electrode layers 140.

5 In all of the illustrated embodiments 3A through 3D, porous electrode layers 140 extend to the edge of ion exchange membrane 130. That is, the electrode layers 140 and the ion exchange membrane 130 are coextensive. The multi-layer MEA 100 may be assembled and then cut to the desired shape and dimensions; then the sealant material 125 may be impregnated into a portion 150 of the porous electrode layers 140. Alternatively, sealant 10 material 125 can be impregnated into a sheet of MEA material. The integral seals for a plurality of MEAs could be injection molded onto the sheet of MEA material, impregnating a plurality of sealing regions of the porous electrode layers 140. After sealant material 125 has cured, the MEA 100 and sealant material 125 may both be cut (preferably in the sealing regions) to the desired dimensions at the same time. Because the sealant material was 15 injection molded prior to the ion exchange membrane being cut, the two electrode layers are kept apart while the sealant material is being injected. Thus the electrode material in the sealing regions is embedded within the electrically insulating sealant material. Cutting the multi-layer material in the sealing regions after the sealant material cures, helps to prevent the possibility of short-circuiting because the cured sealant material immobilizes the 20 embedded electrode material.

25 In the embodiment of FIG. 3A, integral seal 110 extends only as far as the edge of ion exchange membrane 130. That is, the edge of seal 110 is flush with the edge of membrane 130 and electrode layers 140. Therefore, the embodiment shown in FIG. 3A may be made by applying the sealant material before or after the MEA 100 is cut to the desired size and shape.

FIG. 3B illustrates a preferred embodiment. Similar to the embodiment of FIG. 3A, ion exchange membrane 130 is coextensive with porous electrode layers 140 and sealant material is impregnated into a portion 150 of porous electrode layers 140. Unlike the

embodiment of FIG. 3A, sealant material 125 extends laterally beyond the edge of MEA 100, enveloping the edge of ion exchange membrane 130. By enveloping the edge, the sealant material 125 contacts three surfaces of ion exchange membrane 130, namely portions of the two surfaces that face the two electrodes 140 and the side edge defined by 5 the thickness of membrane 130. Integral seal 110 has a single raised rib 160, in the region of the seal that extends beyond the membrane. FIG. 3B also shows an alignment feature in the form of a cylindrical plug or pin 162. Sealant material may be used to make plug 162 that may be molded and formed at the same time as integral seal 110. Plug 162 can cooperate with a corresponding cylindrical depression or well in the adjacent separator plate 10 of a fuel cell to facilitate alignment of MEA 100 with the separator plates during assembly of the fuel cell.

FIG. 3C illustrates an embodiment of an integral seal 110 that has some of the same features as the embodiment depicted by FIG. 3B. However, instead of having only one rib on each face, the embodiment of FIG. 3C has three spaced ribs 165, 170, 175 and cross-ribs 15 180. Those skilled in the art will appreciate that additional ribs will increase the protection against leaks. A breach in one of the ribs will not result in a leak unless there are also breaches in the other parallel ribs. The benefit of the plurality of ribs is augmented by the cross-ribs 180 that compartmentalize the spaces between parallel ribs 165, 170, 175. With the compartmentalized spaces, a leak will not occur unless there is a breach in all three of 20 ribs 165, 170, and 175 within the same compartment between a pair of spaced cross-ribs 180.

FIG. 3D illustrates a preferred embodiment of an integral seal 110 that shares some of the same features as the embodiments depicted by FIGS. 3B and 3C. However, one of the pairs of raised ribs is located in the sealing region that overlaps an edge portion of 25 electrode layers 140 and superposes the membrane. An advantage of this embodiment is that the mechanical pressure that compresses ribs 190 exerts a pinching force on membrane layer 130 to help prevent fluid leaks around the edge of membrane layer 130. The embodiment of FIG. 3D may also employ cross-ribs 195 for further protection against fluid

leaks. FIG. 3D also illustrates the feature of a raised reference edge 197, which may be formed from the sealant material. Reference edge 197 may be used to assist with aligning the MEA with the adjacent fuel cell components, which may be shaped to engage with reference edge 197. Alternatively, reference edge 197 may be used during the 5 manufacturing process to seat the MEA against a guide surface of a machine used to assemble the fuel cells.

FIGS. 4A through 4C show an MEA 100 with an integral seal compressed between two fuel cell separator plates 200. Because at least a portion of MEA integral seal 110 is thicker and/or firmer than MEA 100, the compressive forces acting on the fuel cell stack 10 compress seal 110 against active surfaces 260 of separator plates 200. FIG. 4A depicts the MEA of FIG. 3A compressed between two separator plates 200. FIG. 4B depicts the MEA of FIG. 3B compressed between two separator plates 200. FIG. 4B illustrates an embodiment of the invention wherein surface 260 of plate 200 includes a recessed groove 265. An advantage of this arrangement is that the recessed surface is less prone to scoring 15 or other damage that may occur during the manufacturing process when a number of flow field plates 200 may be stacked one on top of the other. FIG. 4C depicts the MEA of FIG. 3C compressed between two flow field plates 200. FIG. 4C shows that the spacing between parallel ribs 165, 170, and 175 is sufficient to accommodate lateral bulging of the ribs under compression and still provide spaces therebetween.

20 FIG. 5 is a partial plan view of an MEA 100 with an integral perimeter edge seal 110 such as that shown in section view by FIG. 3C. Three spaced parallel ribs 165, 170 and 175 circumscribe the active area of MEA 100. Spaced cross-ribs 180 provide fluidly isolated compartments 185 between ribs 165, 170 and 175.

25 As will be apparent to those skilled in the art in the light of the foregoing disclosure, many alterations and modifications are possible in the practice of this invention without departing from the spirit or scope thereof. Accordingly, the scope of the invention is to be construed in accordance with the substance defined by the following claims.

WHAT IS CLAIMED IS:

1. An improved membrane electrode assembly for an electrochemical fuel cell, said membrane electrode assembly comprising:

5 a first porous electrode layer;

 a second porous electrode layer;

 an ion exchange membrane interposed between said first and second porous electrode layers wherein said first and second electrode layers and said membrane are coextensive;

10 a quantity of electrocatalyst disposed at the interface between said ion exchange membrane and said first and second porous electrode layers, said electrocatalyst defining an electrochemically active area; and

 a resilient fluid impermeable seal integral with said membrane electrode assembly comprising a fluid impermeable sealant material impregnated into said

15 first and second porous electrode layers in sealing regions thereof.

2. The membrane electrode assembly of claim 1 wherein said sealing regions comprise regions that circumscribe said electrochemically active area.

20 3. The membrane electrode assembly of claim 2 wherein said sealing regions comprise regions that circumscribe an opening within said membrane electrode assembly.

25 4. The membrane electrode assembly of claim 1 further comprising an alignment feature molded from said sealant material for assisting alignment of the MEA during assembly of said fuel cell.

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5. The membrane electrode assembly of claim 4 wherein said alignment feature is a protrusion extending perpendicular to a face of said membrane electrode assembly, for cooperating with a corresponding depression in an adjacent separator plate of said fuel cell.

5 6. The membrane electrode assembly of claim 1 wherein said sealant material is an electrical insulator and said seal extends laterally beyond said membrane and said electrode layers, thereby enveloping a peripheral region of both first and second electrodes and said ion exchange membrane.

10 7. The membrane electrode assembly of claim 6 wherein an outer edge of said integral seal comprises a reference edge for assisting assembly of said fuel cell.

8. The membrane electrode assembly of claim 1 wherein said sealant material is an elastomeric material.

15 9. The membrane electrode assembly of claim 8 wherein said elastomeric material is a thermosetting liquid injection moldable compound.

20 10. The membrane electrode assembly of claim 9 wherein said elastomeric material is selected from the group consisting of silicones, fluorosilicones, fluoroelastomers, ethylene propylene di-methyl, and natural rubber.

11. The membrane electrode assembly of claim 1 wherein said integral seal further comprises at least one raised rib.

25 12. The membrane electrode assembly of claim 11 wherein said at least one raised rib comprises a plurality of raised ribs.

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13. The membrane electrode assembly of claim 12 wherein said integral seal comprises a plurality of cross-ribs that extend between said plurality of ribs thereby forming compartmentalized seals.

5 14. The membrane electrode assembly of claim 11 wherein said rib comprises a pair of ribs, wherein a first rib is located in said sealing region where said sealant material is impregnated into said first porous electrode layer and a second rib is located in said sealing region where said sealant material is impregnated into said second porous electrode layer such that said first rib is opposite to said second rib.

10

15. A method of making a membrane electrode assembly with resilient integral seals for use in an electrochemical fuel cell, said method comprising the sequential steps of:

- (a) placing said membrane electrode assembly inside a mold;
- (b) introducing a curable flow processable sealant material into said mold;
- 15 (c) directing said sealant material to desired sealing regions of said membrane electrode assembly and impregnating a portion of an electrode layer of said membrane electrode assembly with said sealant material in said sealing regions;
- (d) curing the sealant material to form said resilient integral seal; and
- 20 (e) removing the membrane electrode assembly.

16. The method of claim 15 wherein step (c) is accompanied by the step of applying a vacuum to facilitate the introduction and impregnation of said sealant material.

25 17. The method of claim 15 wherein said curable sealant material is a thermosetting material and said method further comprises applying heat to cure said thermosetting sealant material.

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18. The method of claim 15 wherein step (c) is facilitated by a mold surface that has open channels for directing said curable flow processable sealant material to said sealing regions, wherein said channels also act as molding surfaces for forming ribs or ridges in said integral seal.

5

19. The method of claim 15 further comprising employing a raised dike in said mold wherein said dike impinges upon said membrane electrode assembly to limit the extent to which said sealant material impregnates said electrode layer.

10 20. The method of claim 15 wherein said membrane electrode assembly is cut from a larger sheet of multi-layer material comprising first and second electrode layers with an ion exchange membrane layer interposed therebetween, and said method further comprises first and second sequential preparation steps completed before step (a); said first preparation step comprising cutting said multi-layer material to a shape and size that is desired for said
15 membrane electrode assembly wherein said first and second electrode layers and said ion exchange membrane layer are coextensive; and said second preparation step comprising brushing cut edges of said multi-layer material to remove electrode material that extends laterally beyond cut edges of said membrane.

20 21. The method of claim 20 wherein said brushing step employs mechanical brushing.

22. The method of claim 20 wherein said brushing step employs compressed air to remove said electrode material that extends laterally beyond said cut edges of said membrane.

25

23. A method of making a membrane electrode assembly with resilient integral seals for use in an electrochemical fuel cell, wherein said membrane electrode assembly is made from a multi-layer material comprising an ion exchange membrane interposed between two

electrode layers and an electrocatalyst disposed on interfacing surfaces between said membrane and said first and second electrode layers; said method comprising the following sequential steps:

- (a) impregnating sealing regions of said first and second electrodes with a fluid impermeable curable flow processable sealant material;
- (b) curing said sealant material; and
- (c) cutting through said multi-layer material in said sealing regions.

5 24. The method of claim 23 wherein said sealing regions define a size and shape of said membrane electrode assembly.

10 25. The method of claim 24 wherein said sealing regions further define the size and shape of openings in said membrane electrode assembly.

15 26. The method of claim 23 wherein said sealant material is injected into a mold to impregnate said electrode layers of said multi-layer material.

27. The method of claim 26 wherein said mold provides channels for forming sealing ribs that protrude above the surface of said electrode layers.

20 28. The method of claim 26 further comprising the step of applying a vacuum to the interior of the mold to assist with injecting said sealant material into said mold.

25 29. The method of claim 26 further comprising employing a raised dike in said mold wherein said dike impinges upon said multi-layer material to limit the extent to which said sealant material impregnates said electrode layer.

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30. The method of claim 23 wherein said sealant material is a thermosetting material and said method further comprises the step of applying heat to cure said thermosetting material.

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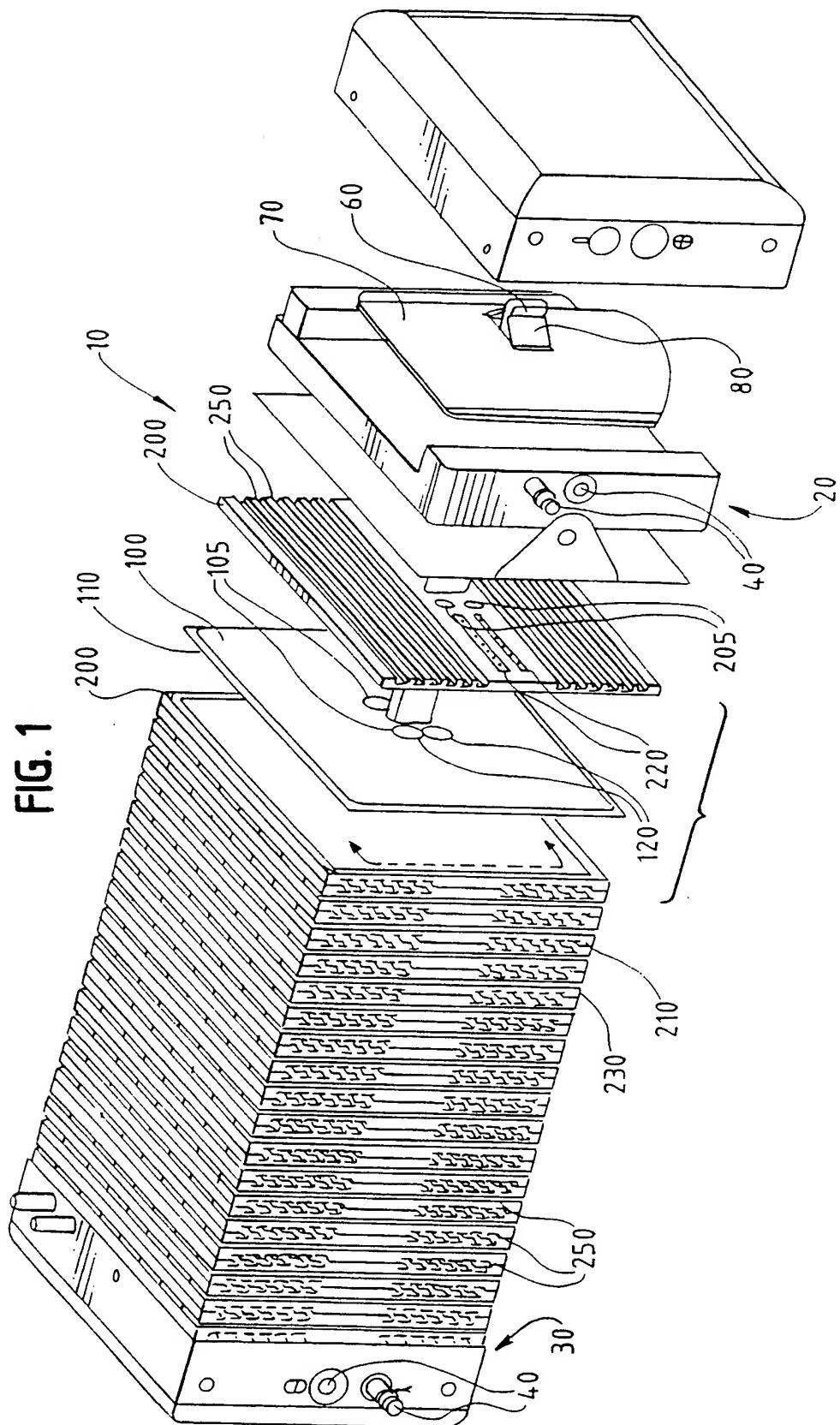
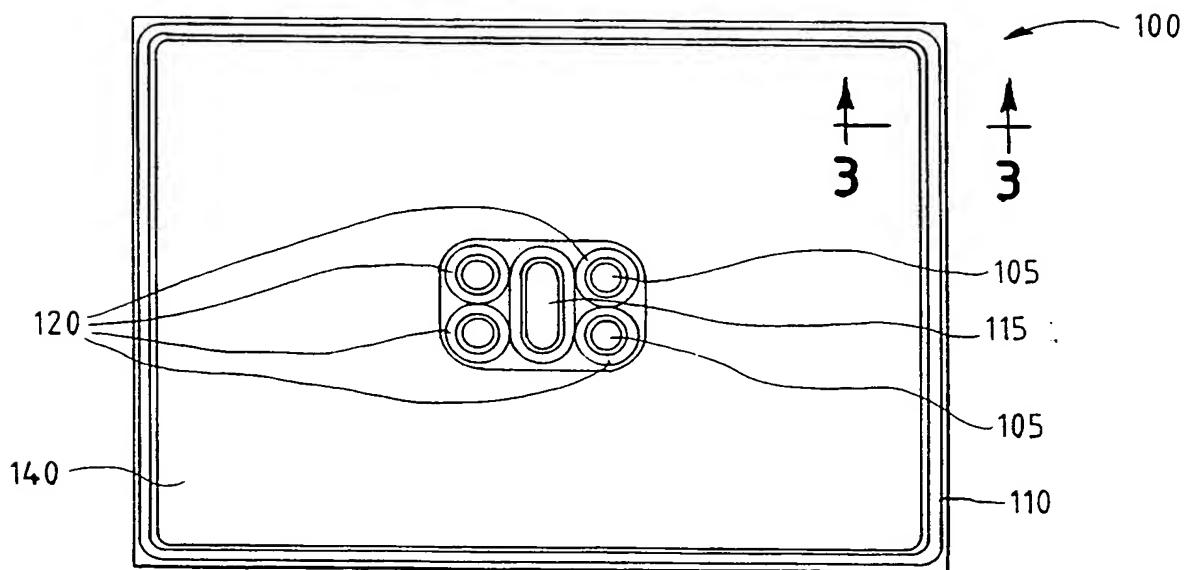


FIG. 1

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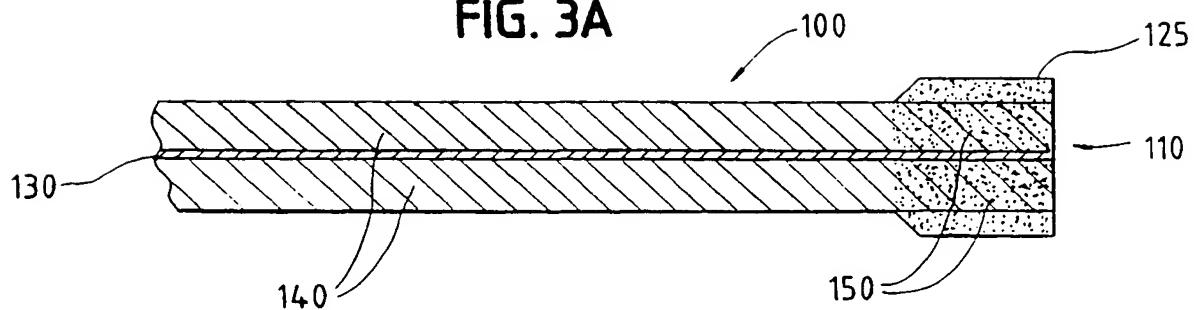
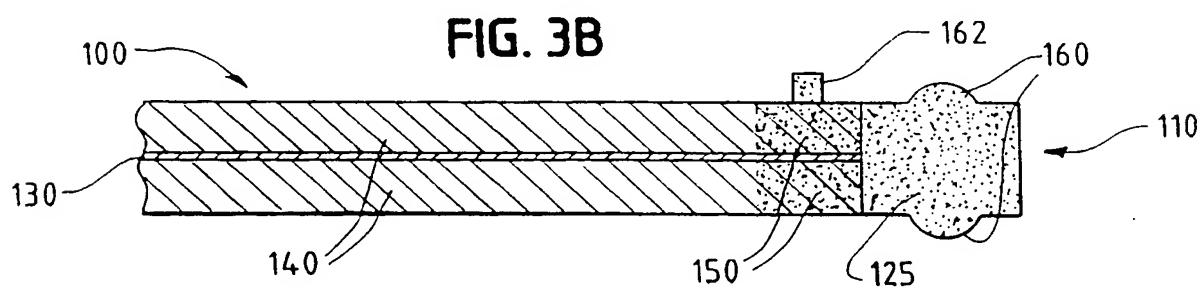
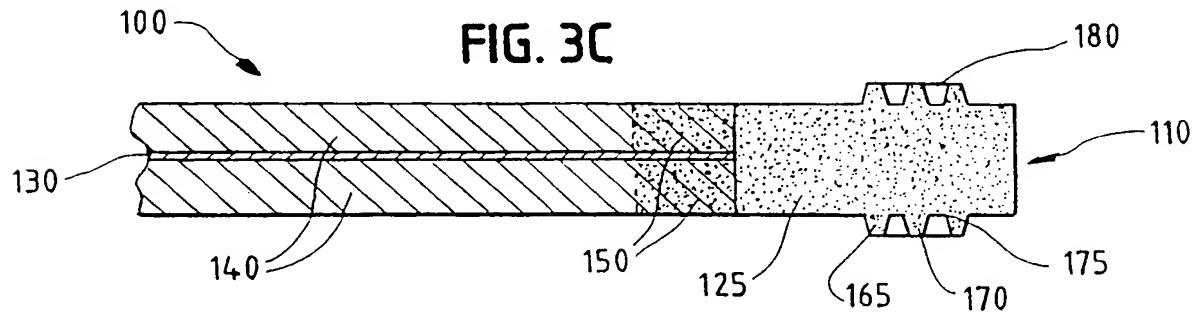
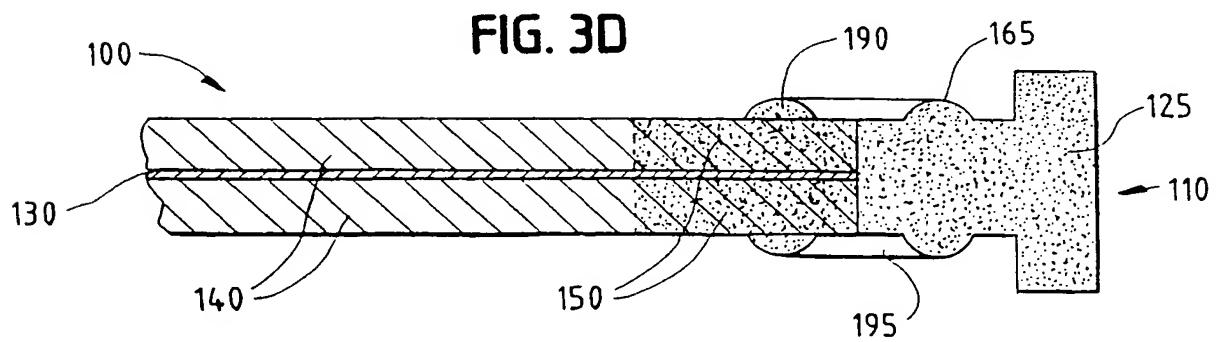
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FIG. 2



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FIG. 3A**FIG. 3B****FIG. 3C****FIG. 3D**

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FIG. 4A

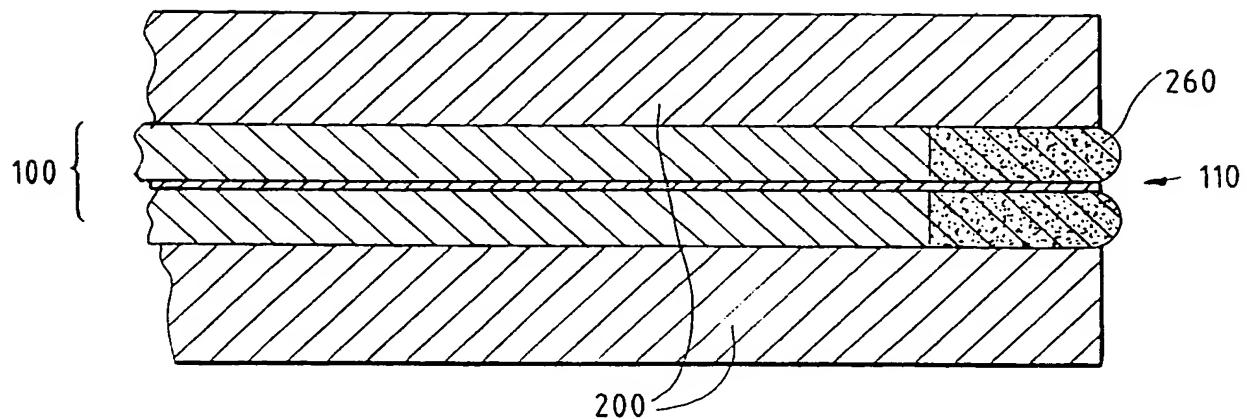


FIG. 4B

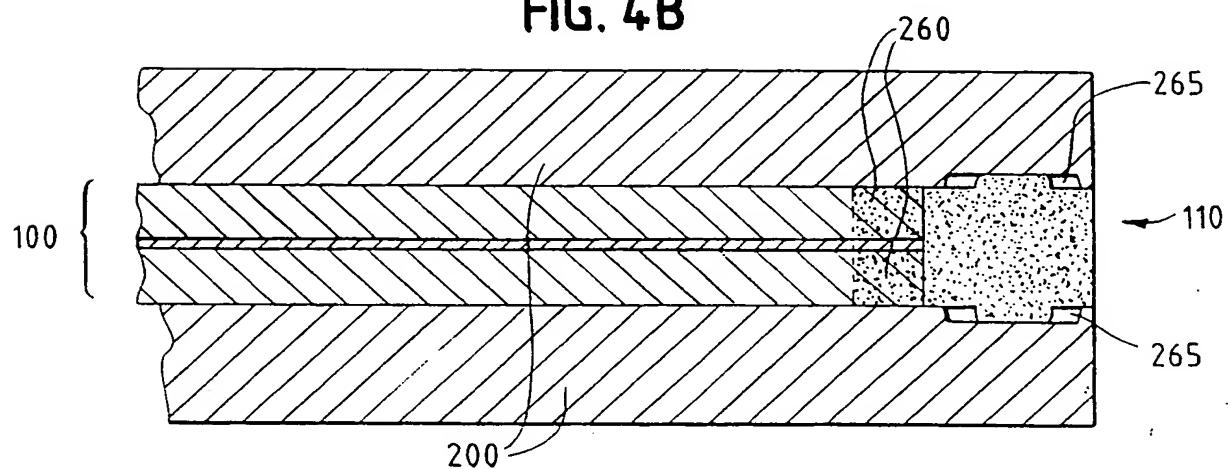
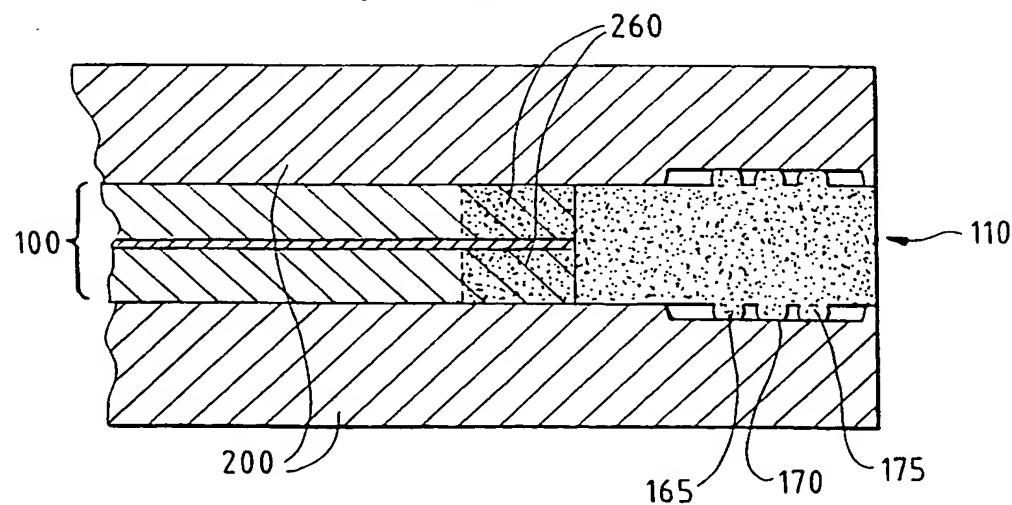


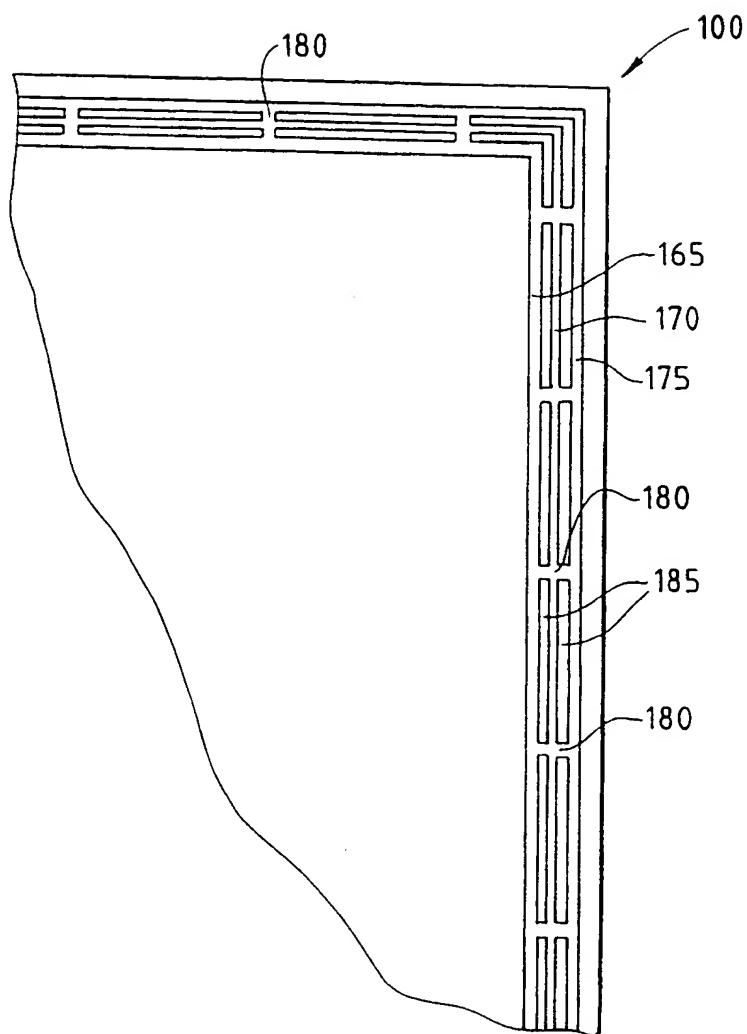
FIG. 4C



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FIG. 5



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INTERNATIONAL SEARCH REPORT

Inte...onal Application No
PCT/CA 98/00687

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H01M8/24 H01M8/02 H01M8/10

According to International Patent Classification(IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|----------|--|-----------------------|
| X | US 5 523 175 A (BEAL DANIEL ET AL) 4 June 1996 | 1-3,8-10 |
| Y | see column 3, line 63 - column 6, line 2 --- | 5,11,12, 14-30 |
| X | EP 0 604 683 A (BALLARD POWER SYSTEMS) 6 July 1994 see column 4, line 29-42 see column 5, line 9-22 | 1-3,8-10 |
| Y | see column 5, line 51 - column 6, line 32 --- | 14-30 |
| X | US 5 219 674 A (SINGELYN JAMES D ET AL) 15 June 1993 see column 2, line 41-63 --- | 1-3,8-10 |
| | | -/- |

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Date of the actual completion of the international search

Date of mailing of the international search report

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18/11/1998

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| C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT | | |
|--|--|-------------------------|
| Category | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
| E | WO 98 33225 A (MAGNET MOTOR GMBH :KOSCHANY ARTHUR (DE); SCHWESINGER THOMAS (DE)) 30 July 1998 see page 14, line 1-14 see page 15, line 3 - page 16, line 7 --- | 1-3, 8-10, 15 |
| Y | WO 92 03854 A (INT FUEL CELLS CORP) 5 March 1992 see claims; figures see page 21, line 22 - page 22, line 2 --- | 1-3, 8-10, 15, 23 |
| Y | US 4 588 661 A (KAUFMAN ARTHUR ET AL) 13 May 1986 see column 2, line 44 - column 3, line 5 see figures --- | 1-3, 8-10, 15, 23 |
| Y | EP 0 774 794 A (DAIMLER BENZ AG) 21 May 1997 see column 3, line 44 - column 5, line 34 see figure 3 ----- | 5, 11, 12, 14 |

INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte onal Application No

PCT/CA 98/00687

| Patent document cited in search report | Publication date | Patent family member(s) | | | Publication date |
|---|---------------------|----------------------------|----|------------|---------------------|
| US 5523175 | A | 04-06-1996 | WO | 9313566 A | 08-07-1993 |
| EP 0604683 | A | 06-07-1994 | | NONE | |
| US 5219674 | A | 15-06-1993 | CA | 2126681 A | 08-07-1993 |
| | | | EP | 0619045 A | 12-10-1994 |
| | | | JP | 7502374 T | 09-03-1995 |
| | | | WO | 9313567 A | 08-07-1993 |
| WO 9833225 | A | 30-07-1998 | AU | 6617298 A | 18-08-1998 |
| WO 9203854 | A | 05-03-1992 | CA | 2090477 A | 01-03-1992 |
| | | | EP | 0546116 A | 16-06-1993 |
| US 4588661 | A | 13-05-1986 | CA | 1250017 A | 14-02-1989 |
| | | | EP | 0174762 A | 19-03-1986 |
| | | | JP | 1939090 C | 09-06-1995 |
| | | | JP | 6065049 B | 22-08-1994 |
| | | | JP | 61061372 A | 29-03-1986 |
| EP 0774794 | A | 21-05-1997 | DE | 19542475 A | 22-05-1997 |

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